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¹ **The Impact of Buoyancy on Front Spreading in**
² **Heterogeneous Porous Media in Two-Phase**
³ **Immiscible Flow**

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4 **Abstract.**

5 We study the influence of buoyancy and spatial heterogeneity on the spread-
6 ing of the saturation front of a displacing fluid during injection into a porous
7 medium saturated with another, immiscible fluid. To do so we use a stochas-
8 tic modeling framework. We derive an effective large-scale flow equation for
9 the saturation of the displacing fluid that is characterized by six nonlocal
10 flux terms, four that resemble dispersive type terms and two that have the
11 appearance of advection terms. From the effective large scale flow equation
12 we derive measures for the spreading of the saturation front. A series of full
13 two phase numerical solutions are conducted to compliment the analytical
14 developments. We find that the interplay between density and heterogene-
15 ity leads to an enhancement of the front spreading on one hand and to a renor-
16 malization of the evolution of the mean front position compared with an equiv-
17 alent homogeneous medium. The quantification of these phenomena plays
18 an important role in several applications, including for example carbon se-
19 questration and enhanced oil recovery .

1. Introduction

20 Capturing the influence of physical heterogeneity on flow and transport in geological
21 media is still one of the great challenges facing us today. Even for linear problems, such
22 as single phase flow and transport many questions remain unanswered and while many
23 have been presented with some success, no single clear model has emerged as capable of
24 capturing all effects of heterogeneity (see [e.g., *Dagan*, 1989; *Gelhar*, 1993; *Neuman and*
25 *Tartakovsky*, 2009]). Similarly, accounting for the influence of buoyancy on single phase
26 flow [e.g., *Henry*, 1964; *Kalejaiye and Cardoso*, 2005; *Huppert and Woods*, 1995; *Dentz*
27 *et al.*, 2006] and transport [e.g., *Graf and Therrien*, 2008; *Bolster et al.*, 2007] in porous
28 media is a challenging problem that has a rich body of work dedicated to it.

29 Many interesting and relevant problems in porous media involve the flow and interaction
30 of two immiscible fluids. Relevant examples that receive much attention include CO₂
31 sequestration [e.g., *Bachu*, 2008; *Bachu and Adams*, 2003; *Bryant et al.*, 2008; *Riaz and*
32 *Tchelepi*, 2008] and enhanced oil recovery [e.g., *Lake*, 1989; *Ferguson et al.*, 2009; *Dong*
33 *et al.*, 2009; *Tokunaga et al.*, 2000]. Accounting for the effects of mobility (viscosity
34 differences between phases) and capillarity introduces significant complexity and results
35 in highly nonlinear and coupled governing equations [e.g., *Binning and Celia*, 1999]. Add
36 to this buoyancy effects when the two phases are of differing density and one has a very
37 interesting and challenging problem (even in the absence of heterogeneity).

38 In this work we focus on the interaction of buoyancy and heterogeneity effects on mul-
39 tiphase flows. To do so, we consider a displacement problem where an invading phase
40 displaces another one as depicted in Figure 1. We neglect the influence of capillarity

41 by using the commonly used Buckley-Leverett approximation, which we discuss in more
42 detail in Section 2. In such a displacement problem there is typically a sharp interface
43 between the invading and displaced phases. Spatial variability in the flow field, induced
44 by heterogeneity, cause this sharp interface to vary in space, which results in spreading of
45 the front. At the same time buoyancy plays its role. In the case of a stable displacement,
46 the spreading ultimately induces lateral pressure gradients that slow down the spreading
47 of the interface. Similarly, an unstable injection will result in greater spreading due to
48 buoyancy. This is illustrated clearly in Figure 1 where the results of three numerical
49 simulations are presented, one with no buoyancy effects (Fig. 1a), one with stabilizing
50 buoyancy (Fig. 1b) and one with destabilizing buoyancy (Fig. 1c).

51 To date, in the field of single phase flows, the approaches to capture the effect of het-
52 erogeneity that have achieved most success are stochastic methods. The theory of such
53 approaches is described extensively in the literature [e.g., *Dagan, 1989; Brenner and Ed-*
54 *wards, 1993; Gelhar, 1993; Rubin, 2003*]. In the context here, if one averages transversely
55 across the transition zones depicted in Figure 1, the resulting transition zone between high
56 and low saturation of the displacing fluid can have the appearance of a dispersive mixing
57 zone. It should of course be noted that this averaged dispersive zone does not represent
58 actual mixing as only spreading occurs. However, for applications where the fluid-fluid
59 interfacial area is important, it is important to have model predictions that quantify the
60 spreading zone.

61 Dispersive transition zones in solute transport problems have typically been character-
62 ized by spatial moments and a wide body of literature exists doing so [e.g., *Aris, 1956;*
63 *Gelhar and Arness, 1983; Dagan, 1989; Kitanidis, 1988; Dentz and Carrera, 2007; Bolster*

64 *et al.*, 2009b]. Similar approaches have been applied to two-phase flow, but most work
65 along these lines has been limited to horizontal displacements that neglect buoyancy ef-
66 fects. *Cvetovic and Dagan* [1996] and *Dagan and Cvetkovic* [1996] applied a Lagrangian
67 perturbation theory approach in order to determine the averaged cumulative recovery of
68 the displacing fluid and the spatial moments of the fluid distribution. They found that the
69 heterogeneities cause a dispersive growth of the second moment. However, they did not
70 quantify it. Similarly *Zhang and Tchelepi* [1999] found a dispersion effect for the immis-
71 cible displacement in the horizontal direction. This dispersion coefficient was calculated
72 semi-analytically by numerical means by *Langlo and Espedal* [1995], who also applied a
73 perturbation theory approach. Their approach was extended by *Neuweiler et al.* [2003] to
74 quantify the dispersion coefficient analytically and later by *Bolster et al.* [2009a] to include
75 temporal fluctuations in the flow field. Within the validity of perturbation theory and in
76 direct analogy to single phase flow, they showed that the dispersive growth for neutrally
77 stable displacement was directly proportional to the variance and the correlation length
78 of the permeability field. As such a natural question arises: given the additional influence
79 of buoyancy, can we anticipate the same behavior?

80 For vertical immiscible displacement in the presence of buoyancy effects we anticipate a
81 similar quasi-dispersive transition zone of the averaged front, which will be augmented or
82 suppressed due to buoyancy. The heterogeneity still leads to fluctuations in the velocity
83 field as illustrated in Figure 1. However, the process will be more complicated and not
84 solely due to the stabilizing and destabilizing processes mentioned above. After all, such
85 stabilization/destabilization effects will occur even for single phase miscible displacement

86 [e.g., *Welty and Gelhar*, 1991; *Kempers and Haas*, 1994]), leading to the question what
87 additional role the multiphase nature of this flow plays?

88 In the absence of buoyancy effects the Buckley Leveret problem is governed by a single
89 dimensionless parameter, which is the viscosity ratio (or ratio of the viscosities of the
90 two phases). This dimensionless number does not depend on any of the parameters
91 associated with the flow or porous medium. This means that while heterogeneity in the
92 porous medium induces fluctuations in the flow field it does not affect the fundamental
93 fluid properties in an equivalent homogeneous medium. Thus, the (mean) front positions
94 obtained from the solutions of the homogeneous and heterogeneous media are identical.

95 On the other hand, when one includes buoyancy effects, a second dimensionless number
96 is necessary to describe the system, namely the gravity number. The gravity number
97 physically reflects the ratio of buoyancy to viscous forces. The buoyancy number (defined
98 formally and discussed further in Section 2) is directly proportional to the permeability
99 of the porous medium. Therefore when the permeability field is heterogeneous in space,
100 so too is the buoyancy number. This means that while the viscosity ratio is insensitive to
101 heterogeneity, the gravity number can potentially vary over orders of magnitude depending
102 on how variable the permeability field is. This raises another important and potentially
103 problematic question: as this system is so inherently nonlinear, does the arithmetic mean
104 (or for that matter any other mean) of the gravity number provide a good representative
105 measure of the behavior of the heterogeneous system?

106 In fact, as the buoyancy number varies in space, in a manner directly proportional
107 to the spatial variations in permeability one might anticipate a local contribution to
108 the dispersion front spreading effect beyond the nonlocal contribution that arises from

109 fluctuations in the velocity field. In this paper we aim to answer the following questions
110 regarding buoyancy influenced multiphase immiscible displacement in a heterogeneous
111 medium.

- 112 1. Can we, using perturbation theory, assess the rate of front spreading that occurs?
- 113 2. What measures of the heterogeneous field (e.g. variance, correlation length) control
114 this spreading? Also, why and how do they?
- 115 3. What influence does the heterogeneity in gravity number have? And does the arith-
116 metic mean of the gravity number represent a mean behavior in the heterogeneous system
117 considering that the problems considered here are highly non-linear?

2. Model

118 The flow of two immiscible fluids in a porous medium can be described by conservation
119 of mass and momentum. Momentum conservation is expressed by the Darcy law, which
120 is

$$\mathbf{q}^{(j)}(\mathbf{x}, t) = -\frac{k(\mathbf{x})k_{rj}(S_j)}{\mu_j} [\nabla p_j(\mathbf{x}, t) + \rho_j g \mathbf{e}_1], \quad (1)$$

121 where $\mathbf{q}^{(j)}(\mathbf{x}, t)$ and $p_j(\mathbf{x}, t)$ are specific discharge and pressure of fluid j , μ_j and ρ_j are
122 viscosity and density of fluid j , $k(\mathbf{x})$ is the intrinsic permeability of the porous medium,
123 $k_{rj}[S_j(\mathbf{x}, t)]$ is the relative permeability of phase j (which depends on saturation). The
124 1-direction of the coordinate system is aligned with negative gravity acceleration as ex-
125 pressed by \mathbf{e}_1 , which denotes the unit-vector in the 1-direction. Mass conservation for
126 each fluid is given by [e.g., *Bear*, 1988]

$$\frac{\partial}{\partial t} \omega \rho_j S_j(\mathbf{x}, t) + \nabla \cdot \rho_j \mathbf{q}^{(j)}(\mathbf{x}, t) = 0. \quad (2)$$

127 We assume here that the medium and the fluid are incompressible so that porosity ω
 128 and density ρ_j of each fluid are constant. The saturations S_j of each fluid sum up to one
 129 and the difference of the pressures in each fluid defines the capillary pressure $p_c(S)$

$$S_{nw} + S_w = 1, \quad p_{nw} - p_w = p_c(S_{nw}), \quad (3)$$

130 where $j = nw$ indicates the non-wetting fluid and $j = w$ the wetting fluid. In the problem
 131 studied here we will use two phases $j = i, d$, where i refers to an injected phase and d to a
 132 displaced phase. From here on, S refers to the saturation of the injected phase S_i . From
 133 the incompressibility conditions and mass conservation, it follows that the divergence of
 134 the total specific discharge $\mathbf{Q}(\mathbf{x}, t) = \mathbf{q}^{(i)}(\mathbf{x}, t) + \mathbf{q}^{(d)}(\mathbf{x}, t)$ is zero

$$\nabla \cdot \mathbf{Q}(\mathbf{x}, t) = 0. \quad (4)$$

135 Eliminating $\mathbf{q}^{(i)}(\mathbf{x}, t)$ from Eq. (2) in favor of $\mathbf{Q}(\mathbf{x}, t)$, one obtains [Bear, 1988]

$$\frac{\partial S}{\partial t} + \nabla \cdot \left[\mathbf{Q}f(S) + \frac{k\Delta\rho g}{\mu_d} \mathbf{e}_1 g(S) \right] - \nabla \cdot \left[f(S)k \frac{k_{rd}(S)}{\mu_d} \frac{dp_c(S)}{dS} \nabla S \right] = 0, \quad (5)$$

136 where $\Delta\rho = \rho_d - \rho_i$. We set $\omega = 1$ for simplicity (which is equivalent to rescaling time).
 137 The fractional flow function $f(S)$ and modified fractional flow function $g(S)$ are defined
 138 by

$$f(S) = \frac{k_{ri}(S)}{k_{ri}(S) + Mk_{rd}(S)}, \quad g(S) = k_{rd}f(S). \quad (6)$$

139 where the viscosity ratio M is defined by

$$M = \frac{\mu_i}{\mu_d}. \quad (7)$$

140 In this work we consider the commonly studied problem of one fluid displacing another
 141 immiscible one. We focus on fluid movement in a vertical two-dimensional porous medium
 142 which is initially filled with fluid d . As outlined above, the 1-axis points upwards. Fluid

143 i is injected along a horizontal line at a constant volumetric flux \bar{Q} , displacing fluid d .
 144 We consider flow far away from the domain boundaries and thus disregard boundary
 145 effects. The resulting mean pressure gradient is then aligned with the 1-direction of the
 146 coordinate system. We restrict our focus on flows where capillary pressure effects are
 147 small and thus we neglect them. The approximation to neglect capillary forces implies
 148 thus displacement processes on large length scales, such as that of an oil reservoir, are
 149 considered and that the flow rates are high. The approximation neglects the influence
 150 of small scale heterogeneity of the capillary entry pressure (e.g. *Neuweiler et al.* [2010]).
 151 This might be questionable if residual saturations and macroscopic trapping would be
 152 important. However, as the focus of this paper is the spreading of immiscible displacement
 153 fronts in geotechnical applications, we proceed by neglecting these effects. This problem of
 154 immiscible two phase viscous dominated flow is commonly known as the Buckley-Leverett
 155 problem. Unlike many previous studies we include the influence of buoyancy.

156 We define the dimensionless coordinates, time and total flow by

$$x_i = l\tilde{x}_i, \quad t = \tau_Q\tilde{t}, \quad \mathbf{Q} = \tilde{\mathbf{Q}}\bar{Q}, \quad (8)$$

157 where l is a characteristic length scale such as the length of the domain and the advection
 158 scale τ_Q is defined by $\tau_Q = l/\bar{Q}$. In the following l will be set equal to the correlation scale
 159 of the permeability field $k(\mathbf{x})$. The governing equation reads in non-dimensional terms as

$$\frac{\partial S}{\partial \tilde{t}} + \tilde{\nabla} \cdot \tilde{\mathbf{Q}}f(S) + \frac{\partial}{\partial x_1}Ng(S), = 0, \quad (9)$$

160 where we disregard the capillary diffusion term, conform with the Buckley-Leverett ap-
 161 proximation. We define the (dimensionless) gravity number N by

$$N = \frac{k\Delta\rho g}{\mu_d \bar{Q}}. \quad (10)$$

163 It compares buoyancy forces to forces driving the movement of the front. A positive gravity
 164 number implies a less dense fluid displacing a denser one, a negative gravity number vice
 165 versa. Note that the gravity number is spatially variable because the permeability k is
 166 spatially variable. For convenience, in the following the tildes will be dropped and all
 167 quantities are understood to be dimensionless.

3. Homogeneous Solution

168 In order to study the heterogeneous problem it is important to explore and understand
 169 the homogeneous one, that is, for constant permeability, $k = \text{constant}$. In this case,
 170 Eq. (9) simplifies to

$$\frac{\partial S_h}{\partial t} + \frac{\partial f}{\partial x_1} + N \frac{\partial g}{\partial x_1} = 0. \quad (11)$$

171 where S_h is the homogeneous saturation. The solution of this problem is governed by two
 172 dimensionless quantities, namely the viscosity ratio M and the gravity number N . Both
 173 these numbers determine the form of the solution of (11). Eq. (11) can be solved using
 174 the method of characteristics [e.g., *Marle*, 1981]. The velocity of the characteristics of
 175 constant saturation are given by the derivatives of the total fractional flow function $\phi(S)$

$$\phi(S_h) = f(S_h) + Ng(S_h). \quad (12)$$

176 Owing to the hyperbolic nature of equation (11) the solution has a sharp front that travels
 177 with the front velocity Q^f . It can be written in the scaling form

$$S_h(x_1/t) = S_h^r(x_1/t) H\left(1 - \frac{x_1}{Q^f t}\right) \quad (13)$$

178 where $H(x)$ is the Heaviside step function. The front position is given by $x_f(t) = Q^f t$.

179 The front velocity is

$$Q^f = \frac{d\phi(S_h^f)}{dS_h^f}, \quad (14)$$

180 where the front saturation S_h^f can be determined by the Welge tangent method [e.g.,
181 *Marle*, 1981], which states that

$$\frac{d\phi(S_h^f)}{dS_h^f} = \frac{\phi(S_h^f)}{S_h^f}. \quad (15)$$

182 This implies together with (14) that the front velocity is given by $Q^f = \phi(S_h^f)/S_h^f$.

183 The form of the rear saturation S^r is obtained by the method of characteristics. As
184 outlined above, the characteristic velocities behind the front are given by $d\phi(S_h^r)/dS_h^r$. As
185 iso-saturation points travel with constant velocity, the characteristic velocity at a given
186 point x_1 and time t is

$$\frac{x_1}{t} = \frac{d\phi(S_h^r)}{dS_h^r}. \quad (16)$$

187 The rear saturation is obtained by inverting this relation.

3.1. Homogeneous saturation profiles

188 For negative gravity numbers, when the density of the injected phase is greater than
189 that of the displaced phase, the total fractional flow function ϕ may not be a monoton-
190 ically increasing function and may have a maximum between the front and maximum
191 saturations. This causes the derivative $d\phi(S^h)/dS^h$ to be negative for saturations larger
192 than the saturation at which $\phi(S^h)$ is maximum. As $d\phi(S^h)/dS^h$ is the velocity at which
193 zones of saturation S^h move, this implies that saturation values larger than the value at
194 which velocities turn negative would move in the direction opposite to the flow direction.

195 In order to deal with these unphysical characteristics, a procedure similar to the one to

196 determine the position of the shock front exists [e.g., *Lake*, 1989]. It results in saturation
197 distributions that are either constant at a value smaller than one until the abrupt front
198 position, or are constant until they reach a transition zone in which saturation decreases
199 to the front value.

200 This behavior reflects the fact that buoyancy carries the injected phase away too quickly
201 for the medium to saturate. Thus, the saturation close to the injection boundary is always
202 smaller than one and remains at this value up to a certain point that is determined by the
203 injection rate and buoyancy. This is illustrated in Fig. 2 for a gravity number of $N = 5$.

204 In order to illustrate the influence of the dimensionless numbers M and N on the ho-
205 mogeneous solutions a sample set is illustrated in Figure 2. All solutions are for quadratic
206 functions as relative permeabilities. In Figure 2a we see the influence of varying N while
207 maintaining M constant. Decreasing N increases the value of the front saturation. This
208 is because buoyancy pulls back the advancing intruding phase thus causing higher local
209 saturations. As the area under all the curves must be the same due to mass conservation
210 the larger the gravity number the further into the domain the injected phase will intrude.
211 Similarly, Fig. 2b illustrates the influence of varying M while maintaining constant N .
212 Decreasing this viscosity ratio decreases the value of the front saturation, causing deeper
213 intrusion of the displacing phase. This is a reflection of the fact that the less the viscosity
214 of the displacing phase, the easier it is for this phase to slip through the porous matrix.
215 This mechanism, whereby it is easier for the invading fluid to slip through the porous ma-
216 trix, can lead to instabilities in the interface that lead to fingering patterns [e.g. *Saffman*
217 *and Taylor*, 1958]. Buoyancy, if the invading phase is less dense than the displaced on,

218 can similarly induce gravitational instabilities [e.g. *Noetinger et al.*, 2004]. A criterion for
 219 these instabilities is outlined in section 3.2

220 The location of the front may be analyzed by looking at the derivative of the saturation
 221 field as this has a sharp delta function at the front, which allows the quantification of
 222 spreading around it [*Bolster et al.*, 2009a]. The expression for the derivative of saturation
 223 is given by

$$224 \quad -\frac{\partial S_h}{\partial x_1} = -\frac{\partial S_h^r(x_1/t)}{\partial x_1} H\left(\frac{x_1}{Q^f t} - 1\right) + \frac{1}{Q^f t} S^r(x_1/t) \delta\left(1 - \frac{x_1}{Q^f t}\right). \quad (17)$$

225 The derivatives of saturation for the profiles in Figure 2b are shown in Figure 3. Here
 226 the delta function at the front is clearly illustrated.

3.2. Stability of the solution

227 The solution of (11) can become unstable. Both viscous and gravity forces have an
 228 impact on the stability of the solution. If the total viscosity ($k_{\text{rel},1}/\mu_1 + k_{\text{rel},2}/\mu_2$) directly
 229 behind of the front is greater than the total viscosity directly ahead of the front the
 230 interface becomes unstable [e.g., *Saffman and Taylor*, 1958; *Riaz and Tchelepi*, 2006].

231 On the other hand, for $\Delta\rho < 0$ gravity tends to damp out perturbations to the interface
 232 if the displacing fluid is heavier than the displaced fluid. Conversely if $\Delta\rho > 0$ any
 233 perturbation will be enhanced. A criterion for stability can be found by introducing a
 234 critical velocity [*Noetinger et al.*, 2004]

$$q_{\text{crit}} = \frac{k S^f \Delta\rho g}{\mu_d (M_{\text{shock}}(S^f)^{-1} - 1)}, \quad (18)$$

235 where

$$M_{\text{shock}}(S) = \frac{(k_{ri}/\mu_i + k_{rd}/\mu_d)|_{S=S^f}}{(k_{ri}/\mu_i + k_{rd}/\mu_d)|_{S=0}}. \quad (19)$$

236 Solutions with flow velocities q_{total} will be stable if

$$q_{total} - q_{crit} < \frac{M_{shock}}{M_{shock} - 1} \quad (20)$$

237 and unstable otherwise. In a heterogeneous medium the heterogeneities cause perturba-
238 tions of the interface between the fluids. Depending on the stability criteria of the flow
239 these perturbations can either be enforced or damped out. Thus heterogeneities can either
240 trigger fingering or be counteracted if the flow is stabilizing.

4. Large Scale Flow Model

241 In the following, we derive large scale flow equations by stochastic averaging of the
242 original local scale flow equation. This results in a large scale effective flow equation for
243 the average saturation. In section 5, using this effective flow equation, we define measures
244 for the front spreading due to fluctuations in the permeability field.

4.1. Stochastic Model

245 We employ a stochastic modeling approach in order to quantify the impact of medium
246 heterogeneity on the saturation front of the displacing fluid. The spatial variability of
247 the intrinsic permeability $k(\mathbf{x})$ is modeled as a stationary correlated stochastic process
248 in space. Its constant mean value is $\overline{k(\mathbf{x})} = \bar{k}$, where the overbar denotes the ensemble
249 average. We decompose the permeability into its mean and (normalized) fluctuations
250 about it,

$$k(\mathbf{x}) = \bar{k} [1 + \kappa(\mathbf{x})] \quad (21)$$

251 Their correlation function of the permeability fluctuations is

$$\overline{\kappa(\mathbf{x})\kappa(\mathbf{x}')} = C^{kk}(\mathbf{x} - \mathbf{x}'). \quad (22)$$

252 The variance and correlation length are defined by

$$\sigma_{kk}^2 = C^{kk}(\mathbf{0}), \quad l_{kk}^2 = \frac{\int d^2x C^{kk}(\mathbf{x})}{\sigma_{kk}^2}. \quad (23)$$

253 For simplicity, we assume the permeability is statistically isotropic. The gravity
 254 number (10) is a linear function of permeability. Using the decomposition (21), it is given
 255 by

$$N(\mathbf{x}) = \overline{N} [1 + \kappa(\mathbf{x})] \quad (24)$$

256 where the mean gravity number is given by

$$\overline{N} = \frac{\overline{k} \Delta \rho g}{\mu_d \overline{Q}}. \quad (25)$$

257 We consider injection of the displacing fluid at an injection plane perpendicular to the
 258 one-direction of the coordinate system. The boundary flux in dimensionless notation is
 259 equal to $\overline{Q} = 1$. The spatial randomness is mapped onto the phase discharges and thus
 260 on the total discharge via the Darcy equations (1), which renders the total discharge a
 261 spatial random field as well. Due to the boundary conditions the (dimensionless) mean
 262 flow velocity is $\overline{\mathbf{Q}(\mathbf{x}, t)} = \mathbf{e}_1$. Thus, we can decompose the total flux into its (constant)
 263 mean value and fluctuations about it

$$\mathbf{Q}(\mathbf{x}, t) = \mathbf{e}_1 + \mathbf{q}'(\mathbf{x}). \quad (26)$$

264 Note that $\mathbf{q}'(\mathbf{x}, t)$ in principle depends on saturation. However, since it is driven by a
 265 constant boundary flux, it is a reasonable approach to consider the total flow velocity as
 266 independent of saturation. In particular, it is worth noting that this is a good assumption
 267 away from the front position. This is no longer valid close to the front [e.g., *Neuweiler*
 268 *et al.*, 2003]. Thus, strictly speaking, the velocity fluctuations cannot be considered sta-

269 tionary and thus the velocity correlation function is given by

$$\overline{q'_i(\mathbf{x})q'_j(\mathbf{x}')} = C_{ij}^{qq}(\mathbf{x}, \mathbf{x}'). \quad (27)$$

270 The cross-correlation between the velocity and permeability fluctuations are accordingly

$$\overline{q'_i(\mathbf{x})\kappa(\mathbf{x}')} = C_i^{kq}(\mathbf{x}, \mathbf{x}'). \quad (28)$$

4.2. Average Flow Equation

271 In analogy to solute transport in heterogeneous media [e.g., *Gelhar and Axness*, 1983;
272 *Koch and Brady*, 1987; *Neuman*, 1993; *Cushman et al.*, 1994], the spread of the ensemble
273 averaged saturation front $\overline{S(\mathbf{x}, t)} \equiv \overline{S}(\mathbf{x}, t)$ due to spatial heterogeneity is modeled by a
274 non-Markovian effective equation. Note that the averaging equation is in general non-
275 Markovian [e.g., *Zwanzig*, 1961; *Kubo et al.*, 1991; *Koch and Brady*, 1987; *Cushman et al.*,
276 1994; *Neuman*, 1993], which is expressed by spatio-temporal non-local flux terms. Under
277 certain conditions, these fluxes can be localized.

278 We follow the methodology routinely applied when deriving average dynamics [e.g., *Koch*
279 *and Brady*, 1987; *Neuman*, 1993; *Cushman et al.*, 1994; *Tartakovsky and Neuman*, 1998],
280 which consists of (i) separating the saturation into mean and fluctuating components,
281 (ii) establishing a (non-closed) system of equations for the average saturation and the
282 saturation fluctuations, (iii) closing the system by disregarding terms that are of higher-
283 order in the variance of the fluctuations of the underlying random fields.

284 Following (24) and (26), we also decompose the saturation into its ensemble mean and
285 fluctuations about it

$$S(\mathbf{x}, t) = \overline{S}(\mathbf{x}, t) + S'(\mathbf{x}, t) \quad (29)$$

286 Assuming that the saturation variance is small we can expand the the fractional flow
 287 function $f(S)$ and $g(S)$ as

$$f(S) = f(\bar{S}) + \frac{\partial f}{\partial S}|_{\bar{S}} S' + \dots, \quad g(S) = g(\bar{S}) + \frac{\partial g}{\partial S}|_{\bar{S}} S' + \dots \quad (30)$$

288 In order to be consistent with the second-order perturbation analysis that follows, the
 289 above expressions should technically be expanded to second order. However, includ-
 290 ing these additional terms significantly complicates the analysis and previous work [e.g.,
 291 *Efendiev and Durlofsky, 2002; Newweiler et al., 2003; Bolster et al., 2009a*] illustrates
 292 that these additional terms do not contribute significantly to the system in the absense
 293 of buoyancy effects. We disregarded them in the following and justify this *a posteriori* by
 294 the agreement with numerical simulations in section 6. The results of this work discussed
 295 in Section 6 also justify this approximation.

296 Using decompositions (24), (26) and (29) as well as (30) in (9), the local scale equation
 297 for the saturation $S(\mathbf{x}, t)$ is given by

$$\begin{aligned} & \frac{\partial \bar{S}(\mathbf{x}, t)}{\partial t} + \frac{\partial S'(\mathbf{x}, t)}{\partial t} + \frac{\partial f(\bar{S})}{\partial x_1} + \frac{\partial}{\partial x_1} \frac{df(\bar{S})}{d\bar{S}} S'(\mathbf{x}, t) + \bar{N} \frac{\partial}{\partial x_1} g(\bar{S}) \\ & + \bar{N} \frac{\partial}{\partial x_1} \frac{dg(\bar{S})}{d\bar{S}} S'(\mathbf{x}, t) + \mathbf{q}'(\mathbf{x}) \cdot \nabla f(\bar{S}) + \bar{N} \frac{\partial}{\partial x_1} \kappa(\mathbf{x}, t) g(\bar{S}) \\ & = -\mathbf{q}'(\mathbf{x}) \cdot \nabla \frac{df(\bar{S})}{d\bar{S}} S'(\mathbf{x}, t) - \bar{N} \frac{\partial}{\partial x_1} \kappa(\mathbf{x}) \frac{dg(\bar{S})}{d\bar{S}} S'(\mathbf{x}, t). \end{aligned} \quad (31)$$

298 Averaging the latter over the ensemble gives

$$\begin{aligned} & \frac{\partial \bar{S}(\mathbf{x}, t)}{\partial t} + \frac{\partial f(\bar{S})}{\partial x_1} + \bar{N} \frac{\partial g(\bar{S})}{\partial x_1} \\ & = -\nabla \cdot \overline{\mathbf{q}'(\mathbf{x}) S'(\mathbf{x}, t)} \frac{df(\bar{S})}{d\bar{S}} - \bar{N} \frac{\partial}{\partial x_1} \overline{\kappa(\mathbf{x}) S'(\mathbf{x}, t)} \frac{dg(\bar{S})}{d\bar{S}}. \end{aligned} \quad (32)$$

299 Subtracting (32) from (31), we obtain an equation for the saturation fluctuations. How-
 300 ever, this system of equations is not closed with respect to the average saturation. In

301 order to close it we disregard terms which are quadratic in the fluctuations obtaining

$$\begin{aligned} \frac{\partial S'(\mathbf{x}, t)}{\partial t} + \frac{\partial}{\partial x_1} \frac{df(\bar{S})}{d\bar{S}} S'(\mathbf{x}, t) + \bar{N} \frac{\partial}{\partial x_1} \frac{dg(\bar{S})}{d\bar{S}} S'(\mathbf{x}, t) \\ = -\mathbf{q}'(\mathbf{x}) \cdot \nabla f(\bar{S}) - \bar{N} \frac{\partial}{\partial x_1} \kappa(\mathbf{x}) g(\bar{S}) \end{aligned} \quad (33)$$

302 This is then solved using the associated Green function, i.e.

$$\begin{aligned} S'(\mathbf{x}, t) = - \int_0^t \int d^d x' G(\mathbf{x}, t | \mathbf{x}', t') \\ \times \left[\mathbf{q}'(\mathbf{x}') \cdot \nabla' f(\bar{S}) + \bar{N} \frac{\partial}{\partial x'_1} \kappa(\mathbf{x}') g(\bar{S}) \right]_{\bar{S}=\bar{S}(\mathbf{x}', t')} \end{aligned} \quad (34)$$

303 where $G(\mathbf{x}, t | \mathbf{x}', t')$ solves

$$\frac{\partial G(\mathbf{x}, t | \mathbf{x}', t')}{\partial t} + \frac{\partial}{\partial x_1} \frac{df(\bar{S})}{d\bar{S}} G(\mathbf{x}, t | \mathbf{x}', t') + \bar{N} \frac{\partial}{\partial x_1} \frac{dg(\bar{S})}{d\bar{S}} G(\mathbf{x}, t | \mathbf{x}', t') = 0 \quad (35)$$

304 for the initial condition $G(\mathbf{x}, t | \mathbf{x}', t') = \delta(\mathbf{x} - \mathbf{x}')$, zero boundary conditions at $x_1 = 0$ and

305 $x_1 = \infty$ and zero normal derivative at the horizontal boundaries. Inserting (34) into (32),

306 we obtain a non-linear upscaled equation for the ensemble averaged saturation

$$\begin{aligned} \frac{\partial \bar{S}(\mathbf{x}, t)}{\partial t} + \frac{\partial f(\bar{S})}{\partial x_1} + \bar{N} \frac{\partial g(\bar{S})}{\partial x_1} \\ - \nabla \cdot \int d\mathbf{x}' \int_0^t dt' \mathcal{A}(\mathbf{x}, t | \mathbf{x}', t') g[\bar{S}(\mathbf{x}', t')] \\ - \nabla \cdot \int d\mathbf{x}' \int_0^t dt' \mathcal{D}^{(g)}(\mathbf{x}, t | \mathbf{x}', t') \nabla' g[\bar{S}(\mathbf{x}', t')] \\ - \nabla \cdot \int d\mathbf{x}' \int_0^t dt' \mathcal{D}^{(f)}(\mathbf{x}, t | \mathbf{x}', t') \nabla' f[\bar{S}(\mathbf{x}', t')] = 0, \end{aligned} \quad (36)$$

307 where the advection kernel $\mathcal{A}(\mathbf{x}, t | \mathbf{x}', t')$ is defined by

$$\begin{aligned} c_i(\mathbf{x}, t | \mathbf{x}', t') = \bar{N} \frac{df[\bar{S}(\mathbf{x}, t)]}{d\bar{S}} G(\mathbf{x}, t | \mathbf{x}', t') \frac{\partial C_i^{kq}(\mathbf{x}, \mathbf{x}')}{\partial x'_1} \\ + \delta_{i1} \bar{N}^2 \frac{dg[\bar{S}(\mathbf{x}, t)]}{d\bar{S}} G(\mathbf{x}, t | \mathbf{x}', t') \frac{\partial C^{kk}(\mathbf{x} - \mathbf{x}')}{\partial x'_1}. \end{aligned} \quad (37a)$$

308 The dispersion kernels have four contributions in total, two of which are due to auto-
 309 correlations of the velocity and permeability fluctuations and two due to cross-correlations
 310 between them,

$$\begin{aligned} \mathcal{D}_{ij}^{(g)}(\mathbf{x}, t|\mathbf{x}', t') &= \delta_{j1} \overline{N} \frac{df[\overline{S}(\mathbf{x}, t)]}{d\overline{S}} G(\mathbf{x}, t|\mathbf{x}', t') C_i^{nq}(\mathbf{x}, \mathbf{x}') \\ &\quad + \delta_{i1} \delta_{j1} \overline{N}^2 \frac{dg[\overline{S}(\mathbf{x}, t)]}{d\overline{S}} G(\mathbf{x}, t|\mathbf{x}', t') C^{mn}(\mathbf{x} - \mathbf{x}') \end{aligned} \quad (37b)$$

$$\begin{aligned} \mathcal{D}_{ij}^{(f)}(\mathbf{x}, t|\mathbf{x}', t') &= \frac{df[\overline{S}(\mathbf{x}, t)]}{d\overline{S}} G(\mathbf{x}, t|\mathbf{x}', t') C_{ij}^{qq}(\mathbf{x}, \mathbf{x}') \\ &\quad + \delta_{i1} \overline{N} \frac{dg[\overline{S}(\mathbf{x}, t)]}{d\overline{S}} G(\mathbf{x}, t|\mathbf{x}', t') C_j^{nq}(\mathbf{x}, \mathbf{x}'). \end{aligned} \quad (37c)$$

311 The first contribution in (37c) quantifies the impact on the large scale flow behavior
 312 due to velocity fluctuations, which has been quantified in *Bolster et al.* [2009a]. The
 313 remaining terms reflect the added influence of buoyancy, which manifest themselves due
 314 to cross-correlation between velocity and permeability fluctuations.

315 Note that equation (36), the large scale flow equation for the mean saturation, has the
 316 structure of a non-linear advection-dispersion equation characterized by spatio-temporal
 317 non-local advective and dispersive fluxes. As outlined above, such non-local fluxes typi-
 318 cally occur when averaging. While in the absence of buoyancy, the spatial heterogeneity
 319 gives rise to a non-linear and non-local dispersive flux, in the presence of buoyancy, there
 320 are additional contributions to this dispersive flux as well as disorder-induced contribu-
 321 tions to the advective flux as quantified by the kernel $\mathcal{A}(\mathbf{x}, t|\mathbf{x}', t')$.

322 Note that the non-linear character of the two-phase problem is preserved during the
 323 upscaling exercise. The non-linearity of the problem is quasi-decoupled in terms of the
 324 Green function; Eq. (35) for $G(\mathbf{x}, t|\mathbf{x}', t')$ is linear but depends on the average saturation.

5. Quantification of Average Front Spreading by Apparent Dispersion

325 In direct analogy to solute transport we will quantify the additional spreading that
326 occurs due to heterogeneity by an apparent dispersion coefficient. It should be stressed
327 that the apparent dispersion coefficient does not only capture effects due to an effective
328 dispersion term in the averaged flow equation (36). The renormalized advective flux term
329 quantified by the kernel (37a) also contributes to the evolution of the apparent dispersion
330 coefficient as defined below.

5.1. Spatial moments

331 As done by *Bolster et al.* [2009a] we will study the influence on the derivative of the
332 saturation, given by

$$\bar{s}(\mathbf{x}, t) = -L^{-1} \frac{\partial \bar{S}(\mathbf{x}, t)}{\partial x_1}. \quad (38)$$

333 where L is the horizontal extension of the flow domain. Recall that fluid is injected
334 over the whole medium cross-section. The motivation for this is that the homogeneous
335 solution develops a shock front, which is captured sharply by measuring the derivative.
336 The resulting averaged profile under the influence of heterogeneity has an appearance
337 similar to a Gaussian type bell that diffuses about this sharp delta function (much like a
338 point injection in the case of single phase solute transport). The goal is to quantify the
339 spreading of the averaged front of $\bar{S}(x, t)$ by the width of the averaged profile of $\bar{s}_i(x, t)$.
340 For an illustration see Figure 9.

341 In analogy to the definition of the width of a tracer plume by spatial moments, we will
342 analyze the spatial moments of $\bar{s}(x, t)$. Let us define the first and second moments in

343 direction of the mean flow by

$$m_1^{(1)}(t) = \int d^2x x_1 s(\mathbf{x}, t), \quad m_{11}^{(2)}(t) = \int d^d x_1^2 s(\mathbf{x}, t), \quad (39)$$

344 The second centered moment

$$\kappa_{11}(t) = m_{11}^{(1)}(t) - m_1^{(2)}(t)^2. \quad (40)$$

345 describes the width of the saturation front. The growth of the width of the saturation
 346 front is characterized by an apparent dispersion, which we define as half the temporal rate
 347 of change of the second centered moment as

$$D^a(t) = \frac{1}{2} \frac{d\kappa_{11}}{dt}. \quad (41)$$

348 Equations for the moments (39) and thus for $D^a(t)$ are derived in Appendix B by invoking
 349 first order perturbation theory.

350 We identify three contributions to $D^a(t)$, i.e.

$$D^a(t) = D^h(t) + D^A(t) + D^e(t). \quad (42)$$

351 $D^h(t)$ is the contribution to spreading that occurs with the rarefaction wave of the
 352 homogeneous solution. $D^A(t)$ are the contributions that occur to the nonlocal advection
 353 kernel \mathcal{A} and $D^e(t)$ those that occur due to the nonlocal dispersive kernels $\mathcal{D}^{(g)}$ and $\mathcal{D}^{(f)}$.

5.2. Homogeneous contribution to spreading

354 The homogeneous contribution $D^h(t)$ is given by

355

$$D^h(t) = \int dx_1 \{f[S_h(x_1/t)] + \overline{N}g[S_h(x_1/t)]\} - t. \quad (43)$$

356 The width of the saturation profile evolves purely due to advective widening as expressed
 357 by the terms $D^h(t)$ and $D^A(t)$ and due to actual front spreading as expressed by $D^e(t)$.

358 For a homogeneous medium, the growth of the width of the saturation profile is due to
359 the fact that different saturations have different characteristic velocities. The term $D^h(t)$
360 is identical to the one that measure this effect in a homogeneous medium [e.g., *Bolster*
361 *et al.*, 2009a]. We can see from (36) that heterogeneity leads to an additional advective
362 flux, which contributes to this purely advective increase of the width of the saturation
363 profile. This is quantified by the term $D^A(t)$. The actual front spreading is quantified by
364 $D^e(t)$. The homogeneous contribution $D^h(t)$ can be obtained by rescaling the integration
365 variable x_1 in (43) according to $x_1 = \eta t$, which gives

$$D^h(t) = t \left\{ \int d\eta \{ f[S_h(\eta)] + \bar{N}g[S_h(\eta)] \} - 1 \right\}. \quad (44)$$

366 Thus, as detailed in [e.g., *Bolster et al.*, 2009a], purely advective effects due to different
367 characteristic velocities lead to a linear evolution of the width of the saturation distribu-
368 tion. Here we observe that for a heavier fluid displacing a lighter one, that is, $\bar{N} < 0$, (25),
369 the increase of the width is slowed down by gravity.

5.3. Contributions from advective kernels to spreading

370 In Appendix B, we derive for the contribution $D^A(t)$ for dimensionless times $t \gg 1$

$$D^A(t) = -t \int_0^\infty d\eta \eta^{-1} \left\{ \bar{N} \sigma_{kq}^2(\eta t) \frac{df[S_h(\eta)]}{dS_h} + \bar{N}^2 \sigma_{kk}^2 \frac{dg[S_h(\eta)]}{dS_h} \right\} g[S_h(\eta)] \\ + \int_0^\infty d\eta \eta^{-2} \left\{ \bar{N} \sigma_{kq}^2(\eta t) l_{kq}(\eta t) \frac{df[S_h(\eta)]}{dS_h} + \bar{N}^2 \sigma_{kk}^2 l_{kk} \frac{dg[S_h(\eta)]}{dS_h} \right\} g[S_h(\eta)]. \quad (45)$$

371 where we defined

$$\sigma_{kq}^2(\eta t) = C_0^{kq}(\eta t, \eta t), \quad \sigma_{kq}^2(\eta t) l_{kq}(\eta t) = \int_0^\infty dx C_0^{kq}(\eta t, x). \quad (46)$$

372 $C_0^{kq}(\eta t, x)$ is defined in (B5). The variance and correlation length of the permeability field
373 are given by (23). They are constant as $k(\mathbf{x})$ is modeled as a stationary random field.

374 Here we identify two contributions, one that evolves linearly with time and a second
 375 contribution that evolves towards a constant value at large times.

5.4. Contributions from dispersive kernels to spreading

376 For the contribution $D^e(t)$, we obtain in Appendix B

$$\begin{aligned}
 D^e(t) = & -\bar{N} \int_0^\infty d\eta \frac{df[S_h(\eta)]}{dS_h} \frac{\partial g[S_h(\eta)]}{\partial \eta} \eta^{-1} \sigma_{kq}^2(\eta t) l_{kq}(\eta t) \\
 & - \bar{N}^2 \int_0^\infty d\eta \frac{dg[S_h(\eta)]}{dS_h} \frac{\partial g[S_h(\eta)]}{\partial \eta} \eta^{-1} \sigma_{kk}^2(\eta t) l_{kk}(\eta t) \\
 & - \bar{N} \int_0^\infty d\eta \frac{dg[S_h(\eta)]}{dS_h} \frac{\partial f[S_h(\eta)]}{\partial \eta} \eta^{-1} \sigma_{kq}^2(\eta t) l_{kq}(\eta t) \\
 & - \int_0^\infty d\eta \frac{df[S_h(\eta)]}{dS_h} \frac{\partial f[S_h(\eta)]}{\partial \eta} \eta^{-1} \sigma_{qq}^2(\eta t) l_{qq}(\eta t). \tag{47}
 \end{aligned}$$

377 The variance and correlation length of the velocity fluctuations are defined as

$$\sigma_{qq}^2(\eta t) = C_0^{qq}(\eta t, \eta t), \quad \sigma_{qq}^2(\eta t) l_{qq}(\eta t) = \int_0^\infty dx C_0^{qq}(\eta t, x). \tag{48}$$

378 $C_0^{qq}(\eta t, x)$ is defined in (B5).

5.5. Approximate solutions of the apparent dispersion coefficients

379 In order to further evaluate $D^A(t)$ and $D^e(t)$ we introduce another approximation (which
 380 we justify a posteriori by comparing the numerical and analytical values). For the case of
 381 the homogeneous Buckley-Leverett flow it is well known that behind the saturation front
 382 the derivative of the fractional flow function $\phi(S_h)$ is given by

$$\frac{d\phi(S_h)}{dS_h} = \frac{x_1}{t}. \tag{49}$$

383 at the rear of the saturation profile, see (16). It is this property which allowed *Neuweiler*
 384 *et al.* [2003] and *Bolster et al.* [2009a] to evaluate their expressions for the dispersion

385 coefficients for the non-buoyant case. Buoyancy complicates things in that the fractional
 386 flow function is given by the sum of $f(S)$ and $Ng(S)$, see (12). Under these conditions,
 387 it is no longer trivial to calculate $\frac{df(S)}{dS}$ and $\frac{dg(S)}{dS}$. However, we do know their values
 388 both at the front as well as the injection boundary. Motivated by the results that emerge
 389 from *Neuweiler et al.* [2003] and *Bolster et al.* [2009a] we assume that these vary linearly
 390 between these two points, that is

$$\frac{df(S_h)}{dS_h} = a_f \frac{x_1}{t}, \quad \frac{dg(S_h)}{dS_h} = a_g \frac{x_1}{t}, \quad (50)$$

391 for $x_1 < Q_f t$. The constants a_f and a_g are the respective slopes of $\frac{df(S_h)}{dS_h}$ and $\frac{dg(S_h)}{dS_h}$.
 392 These are given by calculating the saturation at the front S_h^f from condition (15) and
 393 substituting it into the respective expression for $\frac{df(S_h^f)}{dS_h}$ and $\frac{dg(S_h^f)}{dS}$ for the specific form
 394 of relative permeability chosen. A quick study of these functions reveals that in general
 395 they do not vary linearly. However, as they appear inside of an integral it may provide a
 396 reasonable approximation for quadrature purposes. The numbers a_f and a_g are obtained
 397 by simple interpolation between the the derivatives of $f(S_h)$ and $g(S_h)$ at the front and
 398 at the injection point. Note that a_f is positive while a_g can be positive or negative. The
 399 quality of this approximation (50) is discussed Appendix C.

400 Furthermore we assume that the variances and correlation length in (46) and (48) are
 401 constant, which is a reasonable assumption away from the front [e.g., *Neuweiler et al.*,
 402 2003]. Using these approximations and the fact that S_h is given by (13), that is S_h^r is zero

403 for $x_1 \geq Q^f t$, $D^A(t)$, is given by

$$\begin{aligned}
 D^A(t) = & -t \left(\overline{N} \sigma_{kq}^2 a_f + \overline{N}^2 \sigma_{kk}^2 a_g \right) \int_0^{1/Q^f} d\eta g[S_h^r(\eta)] \\
 & + \left(\overline{N} \sigma_{kq}^2 l_{kq} a_f + \overline{N}^2 \sigma_{kk}^2 l_{kk} a_g \right) \int_0^{1/Q^f} d\eta \frac{g[S_h^r(\eta)]}{\eta}. \tag{51}
 \end{aligned}$$

404 Note that due to the negative sign in front of the first term, this contribution can lead to
 405 a reduction of the linear growth of the saturation distribution. For certain values of the
 406 variance and the gravity number it could lead to negative values for the evolution of the
 407 front width, which is clearly unphysical. This, however, is a relic of low order perturbation
 408 theory.

409 For the contribution $D^e(t)$ these approximations yield

$$D^e(t) = \overline{N} \sigma_{kq}^2 l_{kq} a_g + a_f \sigma_{qq}^2 l_{qq}, \tag{52}$$

411 where we used that $g[S_h^r(\eta)]$ is zero at the injection boundary and at the front, $g[S_h^r(0)] =$
 412 $g[S_h^r(1/Q^f)] = 0$ and that $f[S_h^r(\eta)]$ is one at the injection boundary and zero at the front,
 413 $f[S_h^r(0)] = 1$ and $f[S_h^r(1/Q^f)] = 0$. Note that strictly speaking, all the results are only
 414 valid for small variances of permeability and velocity.

5.6. Apparent Dispersion

415 The contributions to the apparent dispersion coefficients in (51) and (52) illustrate
 416 various interesting features. The contribution (52) and the second term in (51) are similar
 417 to the contributions predicted by *Neuweiler et al.* [2003] and *Bolster et al.* [2009a] for
 418 uniform horizontal flow. These contributions are proportional to the correlation lengths
 419 and variance of the random fields. However, beyond this constant contribution, there

420 is a further contribution that grows linearly in time given by the first terms in (51).
421 Interestingly, this contribution is independent of the correlation length (a result which we
422 test with numerical simulations in the following section).

423 The linearity with the correlation length of the constant contributions is in direct anal-
424 ogy to the effective dispersion coefficient in a solute transport problem, which is identical
425 to the macrodispersion coefficient [e.g., *Gelhar and Axness, 1983*]. The terms that are only
426 proportional to the variance and independent of the correlation length could be interpreted
427 as analogous to an effective permeability in a single phase flow problem, which is also only
428 proportional to the variance and not to the correlation length. The terms proportional to
429 the correlation length can thus be related to an effective dispersion term in the averaged
430 flow equation (36), while the other terms can be related to effective contributions to the
431 gravity term.

432 The contribution that is linear in time in (51) can thus be interpreted as the way that
433 heterogeneity adds contributions to the buoyant counter-flow of the fluids. This shows
434 that the mean gravity number is only a rough measure to estimate the true flow behavior
435 and does not capture this additional influence of heterogeneity.

6. Numerical Simulations

436 In order to test the solutions presented here we also conducted a numerical study of
437 the buoyant Buckley-Leverett problem in a heterogeneous medium. To do this we used
438 an in house finite volume code, which uses an IPES (Implicit in Pressure and Explicit in
439 Saturation) scheme. The details of the algorithm used can be found in *Hasle et al. [2007]*
440 and the setup is the same as that used in *Bolster et al. [2009a]*. The numerical dispersion
441 using this method was generally found to be small compared with the apparent dispersion

442 ($< 10\%$ typically) we calculate. For situations where buoyancy is excessively stabilizing
443 the condition could not be met.

444 For each set of parameters 100 random permeability fields were generated using a ran-
445 dom generator, which is based on a Fourier transform method. Spatially isotropic per-
446 meability fields were generated with a Gaussian distribution, characterized by a relative
447 variance of σ_{kk}^2 and a correlation length of l_{kk} . All simulations were performed using
448 square functions as relative permeability functions, i.e.

449

$$k_{ri}(S) = S^2, \quad k_{rd}(S) = (1 - S)^2. \quad (53)$$

450 Figure 1 shows three sample saturation fields from single realizations using this method-
451 ology. The first corresponds to the case where there is no density difference between the
452 two phases, the second where the injected phase is denser and the third where the injected
453 phase is less dense than the displaced one. This figure clearly illustrates the stabilizing
454 and destabilizing effect that buoyancy has on spreading by heterogeneity.

455 Figure 4 shows the temporal evolution of average saturation profiles (averaged over 100
456 realizations in each case) for three different cases, clearly displaying the dispersive effect
457 that occurs due to heterogeneity. All cases in this figure are stable. However the influence
458 of buoyancy is evident. The case in the middle where the injected phase is very dense leads
459 to much less spreading than the other two cases. As the system becomes less stabilizing
460 the spreading effect becomes more pronounced. In this work we do not present the results
461 of unstable simulations as it is well known that a perturbation approach such as the one
462 developed here can not capture unstable effects [e.g. *Bolster et al.*, 2009a]. Instead we

463 refer the interested reader to works that explore these instabilities [e.g. *Riaz and Tchelepi*,
464 2004, 2007; *Tartakovsky*, 2010].

465 Figures 5 and 6 illustrate a typical measurement of the dispersion coefficient attributed
466 to heterogeneity. In Figure 5 we illustrate the terms $D^h(t)$ (44) for the homogeneous
467 medium and the apparent dispersion coefficient $D^a(t)$ (41). The heterogeneity-induced
468 contributions $D^A(t)$ (45), and $D^e(t)$, (47). are given by the difference of these two lines,
469 which is shown in Figure 6. Note that as predicted by the theory, we have a constant
470 contribution and a contribution that grows linearly in time. To calculate the constant
471 contribution as well as the one that grows linearly in time we perform a best fit of the
472 late time data. The intercept provides the constant contribution, while the slope gives
473 the linear component. The results shown in Figure 6 are normalized by the constant
474 contribution.

6.1. Influence of Variances

475 As mentioned briefly in the previous section the apparent dispersion coefficient in (52)
476 and (51) illustrates various interesting features. For one, it depends proportionally on the
477 variances of the permeability and velocity fields. This suggests that an increase in the
478 variance of the permeability field should lead to a proportional increase in the dispersion
479 coefficient. This means that the constant contribution should be proportionally larger as
480 should the slope of the linear in time contribution (cf. figure 6).

481 Figure 7 illustrates the normalized dispersion coefficient for a sample case with three
482 different variances, namely $\sigma_{kk}^2 = 0.1, 0.5$ and 1 . The dispersion coefficients are normal-
483 ized by the constant value associated with the $\sigma_{kk}^2 = 0.1$ case (i.e. where the fitting line
484 intersects the vertical axis). As is clearly visible the $\sigma_{kk}^2 = 0.5$ and $\sigma_{kk}^2 = 1$ cases have

485 progressively larger values of this constant contribution. Similarly the slope associated
486 with each case is progressively larger thus reflecting the qualitative influence of the vari-
487 ance of the heterogeneity field. Beyond this qualitative agreement between prediction
488 and simulation the quantitative agreement is also good in that the constant contribution
489 $\sigma_{kk}^2 = 0.5$ is roughly 5 (actually 4.78) times larger than the $\sigma_{kk}^2 = 0.1$ and that the $\sigma_{kk}^2 = 1$
490 case is roughly 10 (actually 9.25) times greater. Similarly the slopes are 5 (actually 4.4
491 times) and 10 (actually 8.9 times) times larger. The fact that the disagreement in the
492 slopes is larger than in the intercepts suggests that this measure is more sensitive to the
493 perturbation approximations used here.

6.2. Influence of Correlation Length

494 One of the interesting features of the dispersion coefficients predicted in (52) and (51)
495 is that the constant contributions all depend proportionally on the correlation length,
496 while the terms that grow linearly in time have no dependence on this. In order to test
497 the validity of this prediction we ran a test case with a variance of $\sigma_{kk}^2 = 0.1$ and two
498 different correlation lengths $l_{kk} = 0.25$ and 0.5 . If the qualitative nature of the prediction
499 in (52) and (51) is correct then the only influence on the dispersion coefficient should be an
500 increase in the constant contribution (or graphically an upwards shift in the intersection
501 with the vertical axis), while the slope of the dispersion coefficient against time should
502 remain constant. Figure 8 illustrates the normalized dispersion coefficient for the proposed
503 case for the two different correlation lengths. The dispersion coefficients are normalized
504 by the constant value associated with the $l_{kk} = 0.25$ case. As predicted the intersect is
505 shifted upwards by a factor of roughly 2 (actually 2.11), while the slope remains almost
506 identical (the slope of the larger correlation length case is only 1.07 times greater). This

507 seems to verify the analytical prediction that the correlation length does not influence
508 the terms that grow linearly in time. Some of the good agreement between theory and
509 simulations can be attributed to the fact that the averaging across a wide injection line
510 can smooth out point to point deviations.

511 It should be noted here that this behavior was difficult to observe for values of N close to
512 and smaller than -1 , suggesting that excessive stabilization due to buoyancy invalidate
513 the perturbation approach and analytical deductions made (see for example *Noetinger*
514 *et al.* [2004]).

6.3. Effective Advection

515 As mentioned in Section 2 and performed in the analysis in this work it can be useful to
516 look at the derivative of the saturation field, rather than saturation field to quantify the
517 spreading around the front. This is due to the delta function that coincides with the front
518 location for the homogeneous solution. A figure illustrating this for a set of numerical
519 simulations is shown in Figure 9. The homogeneous solution depicts a relatively sharp
520 front much like the delta function fronts shown in Figure 3 (some differences exist due to
521 unavoidable numerical dispersion and limited spatial resolution). As expected the average
522 heterogeneous solution is more spread out due to the dispersive effects we have discussed
523 so far. However, another interesting feature is visible here. The peak of the spreading
524 front does not coincide with the front for the homogeneous case. This does not occur for
525 situations when the density of both phases is the same (i.e., $N = 0$), where the peak and
526 homogeneous front coincide.

527 This behavior occurs due to the effective advection terms that arise, namely those
528 associated with \mathcal{A} in (36). These terms quantify the shift of the peak and do not quantify

529 actual spreading of the front. Much as the case presented by *Bolster et al.* [2009a] where
530 they illustrated that when not averaged correctly temporal fluctuations may appear to
531 increase spreading, here one must be cautious in interpreting increases in the second
532 centered moment as spreading of the front. After all, the homogeneous solution has a
533 contribution to spreading $D^h(t)$ and these additional effective advection terms merely
534 add to this effect. The actual spreading of the front is only quantified by the constant
535 contributions. This is physically reassuring as otherwise the theory presented here suggests
536 that the apparent dispersion coefficient could grow linearly in time forever, leading to
537 potentially massive spreading zones, despite the stabilizing effect of buoyancy. A physical
538 interpretation of these effective advection terms and the shift in peaks in Figure 9 is given
539 in the following section.

6.4. Qualitative Interpretation of Results and Observations

540 In Figure 9 we clearly see that the spreading does not occur around the sharp front
541 associated with the homogeneous solution associated with the mean permeability. Instead
542 it occurs at some point further ahead of this sharp front. The natural question that arises
543 is why this is so and in order to interpret this we will resort to a qualitative analysis based
544 on averaging several homogeneous solutions. The main issue here is that the governing
545 system of equations are so nonlinear that the mean permeability (or equivalently gravity
546 number) is not representative of the mean behavior of this system.

547 This can be qualitatively interpreted by considering the following simple case. Consider
548 the situation with viscosity ratio $M = 1$ and three homogeneous media with gravity
549 numbers $N = 0, -2.5$ and -5 respectively. The solutions associated with such a system
550 are shown in Figure 10. Although the mean gravity number in this case is -2.5 it is clear

551 from the figure that the mean front location will lie further ahead of the front associated
552 with this case. This is merely a reflection of the fact that the front location does not scale
553 linearly with gravity number. Thus in a system such as the one we consider here where an
554 array of permeabilities exist it is to be expected that the spreading occurs around a front
555 ahead of that associated with the mean permeability. The effective advection terms are
556 merely telling us that the effective permeability of the system and the mean permeability
557 are not one and the same. Note that the same statement would hold if we had expanded
558 the intrinsic permeability around the geometric mean. *Panilov and Floriat* [2004], who
559 studied a similar problem using homogenization, also found that the mean and effective
560 permeability are not the same. However, they claimed that they only expect the two
561 to be different for nonstationary random permeability fields. In this work our fields can
562 be stationary and we still find a discrepancy. The effective advection term could also
563 lead to an effective shape of the gravity function, so that the introduction of an effective
564 permeability would not be sufficient.

7. Conclusions

565 In the introduction to this article we posed a series of questions regarding the influence
566 of buoyancy and heterogeneity on spreading in two phase flow under the Buckley-Leverett
567 approximation. We remind the reader that these were

- 568 1. Can we, using perturbation theory, asses the rate of spreading that occurs?
- 569 2. What measures of the heterogeneous field (e.g. variance, correlation length) control
570 this spreading? Also, why and how do they?

571 3. What influence does the heterogeneity in gravity number have? And does the arith-
572 metic mean of the gravity number represent a mean behavior in the heterogeneous system?

573 The answer to the first question is that following the methodology of *Neuweiler et al.*
574 [2003] and *Bolster et al.* [2009a], where perturbation theory around the mean behavior is
575 employed, we can estimate the apparent dispersion coefficient, which is a measure for the
576 spreading of the front. The dispersion coefficient that arises is more complex than for the
577 case without buoyancy. When we write an effective equation there are now six distinct
578 nonlocal terms that contribute to it. Four of these terms have the appearance of an
579 effective dispersion and the first of these terms is identical to the case without buoyancy.
580 The other two additional terms look more like contributing as effective advections. This
581 is distinctly different from the case with no buoyancy.

582 The answers to the second and the third question are closely related. We explored the
583 different contributions to the front spreading and illustrate that only two of the dispersive
584 nonlocal terms seem to play an important role in spreading of the interface. These terms
585 are proportional to the variance and the correlation length of the heterogeneous fields.
586 The terms that are advective in appearance appear to have no influence on the actual
587 spreading of the front. Instead these terms reflect the location of the front around which
588 spreading occurs. It is proportional to the variance of the heterogeneous fields, but not
589 related to the correlation length. This front is typically further ahead of the front obtained
590 in a homogeneous field with the arithmetic mean of the intrinsic permeability. Thus
591 these terms represent an effective contribution to the gravity term, which might be an
592 effective intrinsic permeability different from the arithmetic mean. This is unexpected
593 according to previous works. As stabilization slows the front down and leads to a more

594 compact saturation profile, the influence of heterogeneity combined with buoyancy is a
 595 diminishing of the stabilization effect on the averaged front. This effect is not captured
 596 by the arithmetic average of the gravity number. The arithmetic mean of the gravity
 597 number does thus not capture the whole flow behavior in a heterogeneous field.

598 Finally, it is remarkable that the time behavior of the different contributions to the
 599 apparent dispersion could be confirmed by numerical simulations even in a quantita-
 600 tive manner, although they are derived from applying linear perturbation theory to a
 601 highly non-linear problem. When carrying out numerical simulations in fields with large
 602 variances, this is no longer true and demonstrates the limitations of the perturbation
 603 approximation used here.

Appendix A: Green Function

604 The Green function for a homogeneous medium satisfies the equation

$$\frac{\partial G_0(x_1, t|x'_1, t')}{\partial t} + \frac{\partial}{\partial x_1} \frac{d\phi[S_h(x_1, t)]}{dS_h} G_0(x_1, t|x'_1, t') = 0 \quad (\text{A1})$$

605 for the initial condition $G_0(x_1, t|x'_1, t') = \delta(x_1 - x'_1)$. Analyzing the homogeneous prob-
 606 lem (11) using the method of characteristics [e.g., *Marle*, 1981], one finds that the deriva-
 607 tive of the total flow function $\phi[S_h(x_1, t)]$ with respect to S_h is the velocity of the char-
 608 acteristic of $S_h(x_1, t)$ at x_1 at time t . The fact the characteristic velocity for a given
 609 saturation is constant, means that the saturation at a given point was transported there
 610 by a constant velocity, which is given by

$$\frac{d\phi[S_h(x_1, t)]}{dS} = \frac{x_1}{t}. \quad (\text{A2})$$

611 This simplifies (A1) to

$$\frac{\partial G_0(x_1, t|x'_1, t')}{\partial t} + \frac{1}{t} \frac{\partial}{\partial x_1} x_1 G_0(x_1, t|x'_1, t') = 0. \quad (\text{A3})$$

612 The latter can be solved by the method of characteristics and gives

$$G_0(x_1, t|x'_1, t') = \frac{1}{t} \delta\left(\frac{x'_1}{t'} - \frac{x_1}{t}\right). \quad (\text{A4})$$

613 which is identical to the one obtained for the homogeneous medium in the absence of
 614 Buoyancy [e.g., *Neuweiler et al.*, 2003; *Bolster et al.*, 2009a]. As the initial condition for
 615 $G(\mathbf{x}, t|\mathbf{x}', t')$ is given by $\delta(\mathbf{x} - \mathbf{x}')$, the zeroth order approximation of the Green function
 616 is given by

$$G(\mathbf{x}, t|\mathbf{x}', t') = G_0(x_1, t|x'_1, t')\delta(x_2 - x'_2) \quad (\text{A5})$$

Appendix B: Spatial Moment Equations and Apparent Dispersion

617 Applying definition (38) to (36) we obtain an equation for $\bar{s}(\mathbf{x}, t)$

$$\begin{aligned} \frac{\partial \bar{s}(\mathbf{x}, t)}{\partial t} &= L^{-1} \frac{\partial^2 f[\bar{S}(\mathbf{x}, t)]}{\partial x_1^2} + L^{-1} \bar{N} \frac{\partial^2 g[\bar{S}(\mathbf{x}, t)]}{\partial x_1^2} \\ &- L^{-1} \frac{\partial}{\partial x_1} \nabla \cdot \int d\mathbf{x}' \int_0^t dt' \mathcal{A}(\mathbf{x}, t|\mathbf{x}', t') g[\bar{S}(\mathbf{x}', t')] \\ &- L^{-1} \frac{\partial}{\partial x_1} \nabla \cdot \int d\mathbf{x}' \int_0^t dt' \mathcal{D}^{(g)}(\mathbf{x}, t|\mathbf{x}', t') \nabla' g[\bar{S}(\mathbf{x}', t')] \\ &- L^{-1} \frac{\partial}{\partial x_1} \nabla \cdot \int d\mathbf{x}' \int_0^t dt' \mathcal{D}^{(f)}(\mathbf{x}, t|\mathbf{x}', t') \nabla' f[\bar{S}(\mathbf{x}', t')] \end{aligned} \quad (\text{B1})$$

618 Approximating $\bar{S}(\mathbf{x}, t)$ by the homogeneous solution $S_h(x_1/t)$, given in (13), and using

619 the Green function (A5) results in

$$\begin{aligned}
\frac{\partial \bar{S}(x_1, t)}{\partial t} &= L^{-1} \frac{\partial^2}{\partial x_1^2} \phi[S_h(x_1/t)] \\
&- L^{-1} \frac{\partial^2}{\partial x_1^2} \int dx'_1 \int_0^t dt' \mathcal{A}_h(x_1, t|x'_1, t') \varphi_g(x'_1/t') \\
&- L^{-1} \frac{\partial^2}{\partial x_1^2} \int dx'_1 \int_0^t dt' \mathcal{D}_h^{(g)}(x_1, t|x'_1, t') \frac{\partial \varphi_g(x'_1/t')}{\partial x'_1} \\
&- L^{-1} \frac{\partial^2}{\partial x_1^2} \int dx'_1 \int_0^t dt' \mathcal{D}_h^{(f)}(x_1, t|x'_1, t') \frac{\partial \varphi_f(x'_1/t')}{\partial x'_1}
\end{aligned} \tag{B2}$$

620 where $\bar{S}(\mathbf{x}, t)$ in this approximation only depends on x_1 , therefore $\bar{S}(\mathbf{x}, t) \equiv \bar{S}(x_1, t)$.

621 Furthermore, the total fractional flow function $\phi(S_h)$ is defined in (12). For convenience,

622 we have defined the functions

$$\varphi_g(x_1/t) = g[S_h(x_1/t)], \quad \varphi_f(x_1/t) = f[S_h(x_1/t)]. \tag{B3}$$

623 using the fact that S_h has the scaling form (13). Furthermore, we define the advection

624 kernel $\mathcal{A}_h(x_1, t|x'_1, t')$ by

625

$$\begin{aligned}
\mathcal{A}_h(x_1, t|x'_1, t') &= \bar{N} \phi_f(x_1/t) \frac{1}{t} \delta\left(\frac{x_1}{t} - \frac{x'_1}{t'}\right) \frac{\partial C_0^{kq}(x_1, x'_1)}{\partial x'_1} \\
&+ \bar{N} \phi_g(x_1/t) \frac{1}{t} \delta\left(\frac{x_1}{t} - \frac{x'_1}{t'}\right) \frac{\partial C_0^{kk}(x_1 - x'_1)}{\partial x'_1}.
\end{aligned} \tag{B4a}$$

626 where we used the explicit form (A4) of the homogeneous Green function. Additionally,

627 we define

$$\phi_f(x_1/t) = \frac{df[S_h(x_1/t)]}{dS_h}, \quad \phi_g(x_1/t) = \frac{dg[S_h(x_1/t)]}{dS_h}. \tag{B4b}$$

628 using again the fact that S_h has the scaling form (13). With all this, the dispersion kernels
 629 are given by

$$\begin{aligned} \mathcal{D}_h^{(g)}(x_1, t|x'_1, t') &= \overline{N}\phi_g(x_1/t)\frac{1}{t}\delta\left(\frac{x_1}{t} - \frac{x'_1}{t'}\right)C_0^{kq}(x_1, x'_1) \\ &\quad + \overline{N}^2\phi_g(x_1/t)\frac{1}{t}\delta\left(\frac{x_1}{t} - \frac{x'_1}{t'}\right)C_0^{kk}(x_1 - x'_1). \end{aligned} \quad (\text{B4c})$$

$$\begin{aligned} \mathcal{D}_h^{(f)}(x_1, t|x'_1, t') &= \overline{N}\phi_f(x_1/t)\frac{1}{t}\delta\left(\frac{x_1}{t} - \frac{x'_1}{t'}\right)C_0^{qq}(x_1, x'_1) \\ &\quad + \phi_f(x_1/t)\frac{1}{t}\delta\left(\frac{x_1}{t} - \frac{x'_1}{t'}\right)C_0^{kq}(x_1, x'_1), \end{aligned} \quad (\text{B4d})$$

630 where we define the correlation function as

$$C_0^{kq}(x_1, x'_1) = C_1^{kq}(\mathbf{x}, \mathbf{x}')|_{x_2=x'_2=0}. \quad (\text{B5})$$

631 $C_0^{qq}(x_1, x'_1)$ and $C_0^{kk}(x_1, x'_1)$ are defined correspondingly.

632 We obtain an expression for the time derivative of $m_1^{(1)}(t)$ by multiplying (B2) by x_1
 633 and subsequent integration over space. This gives

$$\frac{dm_1^{(1)}(t)}{dt} = 1, \quad (\text{B6})$$

634 where we used that $S_h(0, t) = 1$ and the fact that $f(1) = 1$, $f(0) = 0$, $g(0) = g(1) = 0$,
 635 and that $\mathcal{A}_h(x_1, t|x'_1, t')$ is zero at $x_1 = 0$ and $x_1 = \infty$. The evolution equation of the
 636 second moment $m_{11}^{(2)}(t)$ is obtained by multiplying (B2) by x_1 and subsequent integration

$$\begin{aligned}
\frac{dm_{11}^{(2)}(t)}{dt} &= 2 \int dx_1 \phi[S_h(x_1/t)] \\
&\quad - 2 \int dx_1 \int dx'_1 \int_0^t dt' \mathcal{A}_h(x_1, t|x'_1, t') \varphi_g(x'_1/t') \\
&\quad - 2 \int dx_1 \int dx'_1 \int_0^t dt' \mathcal{D}_h^{(g)}(x_1, t|x'_1, t') \frac{\partial \varphi_g(x'_1/t')}{\partial x'_1} \\
&\quad - 2 \int dx_1 \int dx'_1 \int_0^t dt' \mathcal{D}_h^{(f)}(x_1, t|x'_1, t') \frac{\partial \varphi_f(x'_1/t')}{\partial x'_1}. \tag{B7}
\end{aligned}$$

638 Note that the apparent dispersion coefficient (41) is expressed in terms of $m_1^{(1)}(t)$ and
639 $m_{11}^{(2)}(t)$ as

$$D^a(t) = \frac{1}{2} \frac{dm_{11}^{(2)}}{dt} - m_1^{(1)}(t) \frac{dm_1^{(1)}}{dt}. \tag{B8}$$

640 Therefore, combining (B6) and (B7), $D^a(t)$ can be decomposed as in (42) with

641

$$D^h(t) = \int dx_1 \phi[S_h(x_1/t)] - t \tag{B9}$$

$$D^A(t) = - \int dx_1 \int dx'_1 \int_0^t dt' \mathcal{A}_h(x_1, t|x'_1, t') \varphi_g(x'_1/t') \tag{B10}$$

$$\begin{aligned}
D^e(t) &= - \int dx_1 \int dx'_1 \int_0^t dt' \mathcal{D}_h^{(g)}(x_1, t|x'_1, t') \frac{\partial \varphi_g(x'_1/t')}{\partial x'_1} \\
&\quad - \int dx_1 \int dx'_1 \int_0^t dt' \mathcal{D}_h^{(f)}(x_1, t|x'_1, t') \frac{\partial \varphi_f(x'_1/t')}{\partial x'_1}. \tag{B11}
\end{aligned}$$

642 Inserting the kernel $\mathcal{A}_h(t)$ defined by (B4a), we notice that $D^A(t)$, can be written as

$$D^A(t) = \overline{N} M^A \left(\{\phi_f\}, \{C_0^{kq}\}, \{\varphi_g\}, t \right) + \overline{N}^2 M^A \left(\{\phi_g\}, \{C_0^{kk}\}, \{\varphi_g\}, t \right), \tag{B12}$$

643 where the functional $M^A(\{\phi\}, \{C\}, \{\varphi\}, t)$ is defined by

$$M^A(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty dx_1 \int_0^t dt' \int_0^\infty dx'_1 \phi\left(\frac{x_1}{t}\right) \frac{1}{t} \delta\left(\frac{x_1}{t} - \frac{x'_1}{t'}\right) \frac{\partial C(x_1, x'_1)}{\partial x'_1} \varphi(x'_1/t'), \quad (\text{B13})$$

644 We now rescale $x_1 = \eta t$ and $x'_1 = \eta' t'$. This gives

$$M^A(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty d\eta \int_0^t dt' \int_0^\infty d\eta' \phi(\eta) \delta(\eta - \eta') t' C'(\eta t, \eta' t') \varphi(\eta'), \quad (\text{B14})$$

645 where $C'(a, x) = \frac{\partial C(a, x)}{\partial x}$. Executing the η' -integration gives

$$M^A(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty d\eta \int_0^t dt' \phi(\eta) t' C'(\eta t, \eta t') \varphi(\eta). \quad (\text{B15})$$

646 Rescaling time as $t' = x/\eta$, we obtain

$$M^A(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty d\eta \phi(\eta) \varphi(\eta) \eta^{-2} \int_0^{t\eta} dx x C'(\eta t, x). \quad (\text{B16})$$

647 Integration by parts gives

$$M^A(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty d\eta \phi(\eta) \varphi(\eta) \left[\eta^{-1} t C(\eta t, \eta t) + \eta^{-2} \int_0^{t\eta} dx C(\eta t, x) \right]. \quad (\text{B17})$$

648 For dimensionless times $t \gg 1$, we approximate the latter by

$$M^A(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty d\eta \phi(\eta) \varphi(\eta) \left[\eta^{-1} t C(\eta t, \eta t) + \eta^{-2} \int_0^\infty dx C(\eta t, x + \eta t) \right]. \quad (\text{B18})$$

649 Similarly, we observe that $D^e(t)$, (B11), can be written in the unified form

$$D^e(t) = \overline{N} M^e(\{\phi_f\}, \{C_0^{kq}\}, \{\varphi_g\}, t) + \overline{N}^2 M^e(\{\phi_g\}, \{C_0^{kk}\}, \{\varphi_g\}, t) + \overline{N} M^e(\{\phi_g\}, \{C_0^{kq}\}, \{\varphi_f\}, t) + M^e(\{\phi_f\}, \{C_0^{qq}\}, \{\varphi_f\}, t), \quad (\text{B19})$$

650 where the functional $M^e(\{\phi\}, \{C\}, \{\varphi\}, t)$ is defined by

$$M^e(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty dx_1 \int_0^t dt' \int_0^\infty dx'_1 \phi\left(\frac{x_1}{t}\right) \frac{1}{t} \delta\left(\frac{x_1}{t} - \frac{x'_1}{t'}\right) C(x_1, x'_1) \frac{\partial \varphi(x'_1/t')}{\partial x'_1}. \quad (\text{B20})$$

651 Using the same steps that lead to (B16), we obtain

$$M^e(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty d\eta \phi(\eta) \frac{\partial \varphi(\eta)}{\partial \eta} \eta^{-1} \int_0^{t\eta} dx C(\eta t, x). \quad (\text{B21})$$

652 As above, we approximate the latter for times $t \gg 1$ by

$$M^e(\{\phi\}, \{C\}, \{\varphi\}, t) = - \int_0^\infty d\eta \phi(\eta) \frac{\partial \varphi(\eta)}{\partial \eta} \eta^{-1} \int_0^\infty dx C(\eta t, x + \eta t). \quad (\text{B22})$$

Appendix C: Integral Approximations

653 The approximation (50) considerably reduces the complexity of this problem. To illus-
654 trate that this approximation works well we consider the following integrals

$$A_f = \int_0^\infty dx \frac{t}{x} \frac{df[S_h(x/t)]}{dS_h} \frac{df[S_h(x/t)]}{dx} \quad (\text{C1})$$

$$A_g = \int_0^\infty dx \frac{t}{x} \frac{dg[S_h(x/t)]}{dS_h} \frac{df[S_h(x/t)]}{dx} \quad (\text{C2})$$

655 Using the approximation (50) we obtain

$$A_f = -a_f \quad (\text{C3})$$

$$A_g = -a_g \quad (\text{C4})$$

656 These integrals arise naturally if one were to consider a delta correlated permeability
657 field, which can be thought of as a limit of many other correlation functions. Figure 11
658 compares the integrals obtained numerically and calculated by using approximation (50).
659 Figure 11a illustrates A_f . For all values of N and M chosen, the approximation works

660 very well. Similarly Figure 11b shows the numerical evaluation of A_g compared to a_f . The
661 agreement is very good for larger values of M . For small values of M the approximation
662 only seems to work for values of N that are not close to 0.

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